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Relaxation Phenomena and Stability
of Probability Densities

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A characteristic function whose positive time behavior is proportional to a step response function is constructed in such a way that: all its derivatives at $t=0$ are finite; it has the usual exponential decay behavior for intermediate times; it satisfies the Paley-Wiener bound for long times. The constructed characteristic function $C_{\tau_k}(t|0)$ is piece-wise continuous with behavior determined by different exponentials of a monomial function of t, namely $-(|t|/\tau_k)^k$, termed monomial exponentials, on appropriate segments of time. Continuity conditions at joining points provide relations among the τ_k so only one τ_k is an independent parameter. The occurrence of τ_k well within a particular segment in (positive) time determines the monomial exponential that dominates the behavior of $C_{\tau_k}(t|0)$, and the behavior is then called k-dominant. The k-dominance property is discussed for the probability density $P_{\tau_k}(\omega)$ corresponding to $C_{\tau_k}(t|0)$. A formalism is developed in which the probability density for a summand variable in ω-space maintains k-dominant behavior for its corresponding characteristic function. The property of k-dominant stability for probability densities is thereby introduced. At this point the identification of the positive t portion of

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$C_{T_k}(t|0)$ as a step response function is used to make a comparison with a model of relaxation in complex systems which others have called the Ngai model. The latter involves the introduction of interactions that lead to a modification of a constant decay rate for a linear exponential to a time-dependent one appropriate for fractional exponential behavior. Ngai's predicted relation between the two respective relaxation time parameters corresponds here to a continuity condition for $C_{T_k}(t|0)$. k-dominance and, by implication, k-dominant stability are compatible with the Ngai model. The changes in the behavior of $C_{T_k}(t|0)$ and its decay rate may be viewed as temporal analogs of phase transformations caused by the changes in the nature of the interactions in the system. Also compatible with the Ngai model and the concept of k-dominant stability is a cross-over from one-dominant to α-dominant behavior under change of experimental conditions that is actually observed for selected systems.

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RELAXATION PHENOMENA AND STABILITY OF PROBABILITY DENSITIES

I. Introduction

We are interested in the overall time dependence of the relaxation of a perturbed entity to equilibrium. The overall time dependence of the perturbed part of an entity is frequently tracked in terms of the step function response function[1] $\phi(t)$. The function $\phi(t)$ has maximal value at $t=0$, is continuous, and vanishes as $t \to \infty$. The function $\phi(t)$ may be identified as the positive time part of an even function $\Phi(t)$ which may be related[2,3] to the dissipative part of a scalar impulse response function $\chi''(t)$ by

$$\chi''(t) = - \frac{d\Phi(t)}{dt} , \quad -\infty < t < \infty \qquad (1.1)$$

In the following we only consider situations where

$$\lim_{t \to \infty} \chi''(t) = 0 \qquad (1.2)$$

so that the Fourier transform of $\chi''(t)$, namely the dissipative part of the susceptibility $\tilde{\chi}''(\omega)$, has only a continuous spectrum with no singularity at $\omega=0$. Further, since the Fourier transform of $\chi''(t)$ or the dissipative part of the susceptibility has all moments, all the derivatives of $\chi''(t)$ and hence also $\Phi(t)$ at $t=0$ must be finite.

The dissipative impulse response function $\chi''(t)$ may be related by perturbation theoretical developments such as those of Kubo,[2] Case,[3] and Martin[4] to the equilibrium average of a two-time scalar commutator of a configurational coordinate operator $A(t)$,

$$\chi''(t-t') = \frac{1}{i\hbar} \text{Tr}\{\rho_0 [A(t), A(t')]\} \qquad (1.3)$$

Here ρ_0 is the canonical equilibrium density matrix and we have taken $A(t)$ to be the perturbed part only. Then it is clear that $\chi''(t)$ and also $\Phi(t)$ are functions only of a time difference t.

The equation for $\phi(t)$ corresponding to $\chi''(t)$ in Eq. (1.3) is[2,3]

$$\phi(t) = 2 \sum_{m>m'} \frac{W_m - W_m'}{E_{m'} - E_m} \left|\langle m|A|m'\rangle\right|^2 \cos(E_m - E_{m'})t/\hbar \qquad (1.4)$$

Here E_m is an eigenvalue of the unperturbed Hamiltonian, $W_m = \exp(-\beta E_m)/Z$, $Z = \Sigma \exp(-\beta E_m)$ and $\beta = 1/kT$. It is clear that $\phi(t)$ is even in t. In the high temperature limit, equivalent to a classical limit, $\phi(t)$ is just $\beta/2$ times the canonical average of the anticommutator $\{A(t), A(0)\}$ which may be identified with a real autocorrelation function. In any event $\phi(t)$ shares with the auto-correlation function of a weakly stationary process the general properties that it is a two-time function dependent only on time differences, i.e. location invariant, and continuous for all time. We have also in addition that $\phi(t)$ has other temporal behavior determined by the physical properties of $\phi(t)$ mentioned above, viz. it is maximal and differentiable to all orders at $t=0$, and mono-tonically vanishing as $t \to \infty$. Furthermore from the causality of $\phi(t)$, we infer $\tilde{\chi}''(\omega)$ is one of a Hilbert transform pair.[2] Consistency with this latter property of $\tilde{\chi}''(\omega)$ leads us to the requirement that $\phi(t)$ is square integrable for $-\infty < t < \infty$ i.e. $L^2(-\infty, \infty)$. Also from Eq. (1.4) we may verify that since $\phi(t)$ is continuous at $t=0$, it is also continuous at neighboring points, ε, and hence for all t. Moreover if $X(t)$ is an arbitrary function whose Fourier transform exists, then from Eq. (1.4) it follows that

$$\iint X(t)\phi(t-t')X*(t')dtdt' = \sum_{m \neq m'} \left(\frac{W_m - W_m'}{E_{m'} - E_m}\right) \left|\langle m|A|m'\rangle\right|^2 \cdot$$

$$\left| \int X(t) e^{-\frac{i}{\hbar}(E_m - E_{m'})t} \, dt \right|^2 \geq 0 \qquad (1.5)$$

Bochner's theorem[5] may then be used to prove that $\phi(t)$ is proportional to a characteristic function.

We find it convenient to deal with the normalized function

$$C(t|0) \equiv \phi(t)/\phi(0) \qquad (1.6)$$

where we have made the two time nature of $C(t|0)$ explicit. By its definition $C(0|0) = 1$ so that it is in fact a characteristic function and we shall refer to it as such in the following.

A major purpose of this paper is to construct a class of characteristic functions with behavior that is consistent with the mathematical and phenomenological constraints on the step-response function $\phi(t)$. These requirements are satisfied by allowing a continous $C(t|0)$ to be only differentiable almost everywhere. This leads to a piecewise character of $C(t|0)$ which will be discussed in detail in the next section. The resulting construction is a $C(t|0)$ which has the form of appropriate exponentials of monomial functions of t, namely $-(|t|/\tau_k)^k$, termed monomial exponentials, on appropriate non-overlapping segments of time. For example, for k=2 we have a segment of quadratic exponential behavior, etc. Continuity conditions at the joining points of the segments provide relations among the τ_k. It is pointed out that a $C(t|0)$ so defined is in fact the Fourier transform of a probability density.

With a probability density in hand, we turn to another major purpose of the paper in section III and consider the description of a total system built up from subsystems that share the same generic temporal behavior. We find the conditions under which the probability density for the total system also has the same piecewise characteristic function as that of the subsystems. A new type of stability for probability densities is thereby obtained.

In section IV, we compare the general properties of the piecewise $C(t|0)$ for t>0 with the step-response function in a model of relaxation in complex systems previously introduced[6] and discussed[7] by us. The latter in fact shares two important features with the $C(t|0)$ constructed in section III. Namely it

has: (1) a long time fractional exponential regime preceded by a linear exponential regime; and (2) a relationship between the relaxation times or τ-parameters in the two regimes. The model is reviewed briefly as a way to gain insight into the meaning of the piecewise $C(t|0)$ and the new type of stability for probability densities discussed in section IV.

II. Temporal Behavior of the Characteristic Function

We want now to consider the consequences of the various phenomenological and mathematical constraints on the temporal behavior of the characteristic function $C(t|0)$ introduced in section I. The properties already indicated in that section are the general ones of location invariance and continuity, and the more special ones of evenness in t, differentiability to all orders at t=0 (from the existence of all moments of the dissipative part of the susceptibility), maximum value at t=0, and L^2 $(-\infty, \infty)$.

It should be noted that the physics of all our considerations is nonrelativistic so the details of behavior at or near t=0 is not actually included in our formulation. The best we can hope for near t=0 is a self-consistent phenomenology. However, this self-consistency leads to some interesting constraints. From the differentiability property of $C(t|0)$ at t=0, linear (and all other odd) terms in its power series expansion around t=0 must vanish. In particular, $C(t|0)$ cannot then be taken to have a linear exponential form at t=0. An even more stringent condition arises from the fact that all the derivatives of $C(t|0)$ at t=0 must be finite. Further as already noted, from the physical meaning of step response, $C(t|0)$ is also maximal at t=0. A self-consistent (but not unique) choice for $C(t|0)$ at or near t=0 is a quadratic exponential or a Gaussian form centered on t=0.

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For values of t>0, we expect from empirical evidence covering most particular cases that $C(t|0)$ may appear to decay in one of three general forms: (a) quadratic exponential (Gaussian); (b) linear exponential (most usually calculated, and most often observed in simple systems); (c) fractional exponential (for complex systems). Further for very long times, the real $L^2(-\infty,\infty)$ characteristic function considered here is constrained to be consistent with the Paley-Wiener bound[8]

$$\lim_{|t|\to\infty} C(t|0) > \exp-(|t|/\tau_\alpha)^\alpha, \quad 0<\alpha<1 \qquad (2.1)$$

where τ_α is a positive real scale parameter. It may be noted that the Paley-Wiener bound is a fractional exponential so the latter is the _fastest_ allowable decay as $|t|\to\infty$.

Thus for any $C(t|0)$ there are conditions near or at $t=0$ and for very long times that must be satisfied. At intermediate times, there may be physically determined behavior that differs from at least one[9] and possibly both of the behaviors respectively around $t=0$ and for very long times. We now consider a generic $C(t|0)$ that incorporates all these features and, dependent upon the choice of parameterization, can be used to describe any of the observed monomial exponential time behaviors.

We consider $C(t|0)$ to be piecewise continuous with appropriate monomial exponential behavior in given nonoverlapping segments which cover $-\infty<t<\infty$ completely. The conditions around $t=0$ _and_ for a physical decay regime that has a quadratic monomial behavior can be satisfied by taking $C(t|0)$ to have the form $\exp-(t/\tau_2)^2$ in an initial segment pair joined at $t=0$, i.e. for $0<|t|<T_0$. Here τ_2 is a positive real scale parameter and T_0 is also positive real. The possibility of a linear exponential regime that often occurs empirically and is the form obtained in standard decay calculations can be allowed by choosing $C(t|0)$ to have the form $\exp-|t|/\tau_1$ in a second segment pair $T_0<|t|<T_L$. Here

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gain τ_1 is a positive real scale parameter and T_L is positive real. To allow
or complex physical systems that have a fractional exponential decay and for
ong time behavior at the Paley-Wiener bound, $C(t|0)$ is taken to have the form
xp$-(|t|/\tau_\alpha)^\alpha$, $0<\alpha<1$ for $T_L<|t|<\infty$. Again T_L is positive real. The continuity
f $C(t|0)$ is assured by continuity conditions at its joining points $\pm T_0$, $\pm T_L$.
he time behavior of our generic $C(t|0)$ may be summarized as follows.

$$C(t|0, T_0, T_L; \tau_k) = \begin{cases} \exp-(t/\tau_2)^2 \quad, \quad -T_0<t<T_0 \\ \exp-(T_0/\tau_2)^2 = \exp-(T_0/\tau_1) \quad, \quad |t| = T_0 \\ \exp-|t|/\tau_1 \quad, \quad T_0<|t|<T_L \\ \exp-(T_L/\tau_1)=\exp-(T_L/\tau_\alpha)^\alpha \quad, \quad 0<\alpha<1, \quad |t| = T_L \\ \exp-(|t|/\tau_\alpha)^\alpha, \quad 0<\alpha<1, \quad T_L<|t|<\infty \end{cases}$$

$$(2.2)$$

The symbol τ_k in $C(t|0, T_0, T_L; \tau_k)$ represents any of the $k=\alpha$, 1, 2 since
nly one of the τ_k is an independent parameter. From the continuity condi-
ions, it follows that

$$\tau_2^2 = T_0\tau_1 \qquad\qquad (2.3a)$$

$$\tau_\alpha^\alpha = T_L{}^{\alpha-1}\tau_1 \qquad\qquad (2.3b)$$

he choice of τ_k as a label will be made on physical grounds as described in
ore detail below. For the moment we write

$$C(t|0, T_0, T_L; \tau_k) \equiv C_{\tau_k}(t|0) \qquad\qquad (2.4)$$

nd choose τ_k to be that τ_k which falls within the positive t segment with k
xponential behavior. This can happen for only one of the k values. For
xample, if $T_0<\tau_1<T_L$ we would write $C_{\tau_1}(t|0)$ and note that $\tau_2^2 = T_0\tau_1>T_0^2$,
$\tau_\alpha^\alpha = T_L{}^{\alpha-1}\tau_1<T_L^\alpha$ so that τ_2 is not in $[0,T_0]$ and τ_α is not in $[T_L,\infty]$. Similarly
or $C_{\tau_2}(t|0)$, τ_2 is in $[0,T_0]$ but τ_1 is not in $[T_0,T_L]$ and τ_α is not in $[T_L,\infty]$.

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Finally for $C_{\tau_\alpha}(t|0)$, τ_α is in $[T_L,\infty)$ but τ_2 is not in $[0,T_0]$ and τ_1 is not in $[T_0,T_L]$. We shall see that $C_{\tau_k}(t|0)$ is, for <u>centrally</u> placed τ_k on a logarithmic time scale, dominated by its behavior in the region in which τ_k falls. We shall call $C_{\tau_k}(t|0)$ a k-dominant characteristic function in such cases. However, before addressing this particular point, we want to note some general properties of $C_{\tau_k}(t|0)$.

First it is clear that although $C_{\tau_k}(t|0)$ is continuous, it is only piece-wise differentiable with discontinuities in derivative at $t=\pm T_0$, $\pm T_L$. Further, location invariance is maintained by the requirement that a translation of the origin, e.g. by δ, is accompanied by a like translation of time segment end-points e.g. $-T_L \to -T_L+\delta$ and $T_L \to T_L+\delta$, etc. Also we note that $C_{\tau_k}(t|0)$ is the Fourier transform of a probability density. This follows because the nature of the probability distribution function associated with the characteristic function depends on the manner in which the latter behaves as $|t| \to \infty$.[10] If it goes to zero as with our $L^2(-\infty,\infty)$ class of functions, then a probability distribution function is continuous. For completeness it may be noted that if the characteristic function tends to a constant, D, with $0<D<1$, the associated probability distribution function is singular and may be of the form of a Cantor set, for example. If it tends to unity, then the probability distribution ion function is discrete.

We return now to the discussion of the generic character of $C_{\tau_k}(t|0)$ which is readily exhibited by considering three mutually exclusive cases of parameter magnitude: (a) $0 \ll \tau_2 \ll T_0$; (b) $T_0 \ll \tau_1 \ll T_L$; (c) $T_L \ll \tau_\alpha \ll \infty$. Case (a) corresponds to 2-dominant behavior. The major part of the decay of $C_{\tau_2}(t|0)$ appears as a quadratic exponential, and the linear and fractional exponential parts are submerged in a long time tail of small magnitude. On the basis of its direct effect, we could here omit the linear exponential part. However,

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present construction requires the continuity condition Eq. (2.3a) that pro-
es a relation between τ_2 and τ_1. Thus the linear exponential region would
revealed by comparing functional relations for a measured τ_2 with corre-
nding functional relations for a calculated τ_1. Finally, the fractional
onential or slower behavior must be included at long times to meet the con-
ion of the Paley-Wiener bound.

Case (b) corresponds to 1-dominant behavior. The major part of the decay
$C_{\tau_1}(t|0)$ appears as a linear exponential. The latter is of course the most
monly observed behavior in simple systems and is the usual result of theo-
ical calculations based on a random phase approximation or the introduction
a phenomenological friction term. The quadratic exponential segment of
$(t|0)$ is submerged in small deviations at times short compared to τ_1 but
or some equivalent must be included to meet the conditions on $C_{\tau_1}(t|0)$
$t=0$. Also again a fractional exponential or slower time behavior must be
luded to meet the Paley-Wiener bound at long times even though that region
a small magnitude.

Finally, case (c) corresponds to α-dominant behavior. The major part of
decay of $C_{\tau_\alpha}(t|0)$ appears as a fractional exponential. The quadratic expo-
tial segment has an immeasurable effect at short times but it or its equiva-
t is essential to meet conditions at $t=0$. The linear exponential regime
e provides only small deviations that would most probably also not be noted
measurements. Thus again as in case (a) the linear exponential segment
ht at first sight appear to be unnecessary. However, if there is a linear
onential regime, the measured parameter τ_α can be compared to a calculated
by means of the continuity condition Eq. (2.3b). This continuity condition
n then provides an important relation that is subject to verification. In
t, a similar relation between relaxation time parameters respectively in the

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linear and fractional exponential regimes is a key feature of the model of relaxation in complex systems[6,7] to be discussed below in section IV.

We can gain further insight into the time behavior of $C_{\tau_k}(t|0)$ by consideration of the instantaneous decay rate $W_{\tau_k}(t|0)$ which is defined to be

$$
\left.
\begin{aligned}
W_{\tau_k}(t|0) &= -(d\ C_{\tau_k}(t|0)/dt)/C_{\tau_k}(t|0) \\[2mm]
&= -\ d\ \ell n C_{\tau_k}(t|0)/dt
\end{aligned}
\right\} \quad t \neq \pm\ T_Q,\ T_L \qquad (2.4)
$$

We designate $W_{\tau_k}(t|0)$ to be $W_2(t|0)$, $W_1(t|0)$, and $W_\alpha(t|0)$ respectively in the segments where $C_{\tau_k}(t|0)$ has quadratic, linear and fractional exponential behavior. Since $C_{\tau_k}(t|0)$ is even, it is sufficient to consider $t>0$. In the quadratic exponential segment, $W_2(t|0)=2t/\tau_2^2$ which approaches a maximum value $W_2(T_Q|0)=2T_Q/\tau_2^2=2/\tau_1$ as t approaches T_Q. On the other hand $W_1(t|0)=1/\tau_1$ for all $T_Q<t<T_L$ so there is a discontinuity in $W_{\tau_k}(t|0)$ at $t=T_Q$ where it falls to one half its limiting maximum value in the quadratic segment. Similarly there is a discontinuity at $t=T_L$ for $W_\alpha(t|0) = \alpha t^{\alpha-1}/\tau_\alpha^\alpha$ has a maximum limiting value $W_\alpha(T_L|0) = \alpha/\tau_1 < 1/\tau_1$. It is clear that this sequential behavior for $W_{\tau_k}(t|0)$ is independent of the choice of magnitude τ's and so occurs for all possibilities of the k in k-dominance.

The sharpness of the discontinuities in $W_{\tau_k}(t|0)$ is an artifact of our piecewise construction for $C_{\tau_k}(t|0)$. Nevertheless decay rates depend on system interactions so these or perhaps somewhat more smooth behavior can be given a physical meaning. They can be viewed as due to changes in the nature of the interactions in the system at $|t|=T_Q$, T_L. Such changes can be thought of as temporal transformations of the environment in which the relaxation is occurring. These transformations can be viewed as the analogs of thermodynamic phase transformations.

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The $C_{\tau_k}(t|0)$ of Eq. (2.2) could be readily generalized by addition of other monomial exponential intermediate segments or appropriate short time or long time segments. Such a generalization would also involve corresponding additions (and discontinuities) in $W_{\tau_k}(t|0)$. The point we wish to emphasize however is that the generic $C_{\tau_k}(t|0)$ defined in Eq. (2.2) satisfies all the mathematical requirements for, and can provide an adequate description of the most usually phenomenological behaviors of real step-response functions. A simple and useful property of $C_{\tau_k}(t|0)$ so constructed is that it has monomial exponential behavior in all segments. This has provided simple continuity conditions, Eqs. (2.3a,b), and further, provides the basis for consideration in the next section of stability properties for the probability density $P_{\tau_k}(\omega)$ belonging to $C_{\tau_k}(t|0)$.

III. k-dominant Stability for Probability Densities

We turn now to a discussion of the stability properties of $P_{\tau_k}(\omega)$. The probability density $P_{\tau_k}(\omega)$ corresponding to $C_{\tau_k}(t|0)$ is a function of ω as a random variable that incorporates properties that reflect the particular k-dominant and piecewise properties of $C_{\tau_k}(t|0)$.

$$C_{\tau_k}(t|0) = \int_{-\infty}^{\infty} P_{\tau_k}(\omega) \cos\omega t \, d\omega \tag{3.1a}$$

$$P_{\tau_k}(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} C_{\tau_k}(t|0) \cos \omega t \, dt \tag{3.1b}$$

We note that $C_{\tau_k}(t|0)$ remains invariant under scaling of time by a numerical factor. From Eqs. (2.2) and (2.4),

$$C(At|0, AT_Q, AT_L; A\tau_k) \equiv C_{A\tau_k}(At|0) = C_{\tau_k}(t|0) \tag{3.2}$$

This property merely reflects the fact that the choice of scale in temporal measurements does not affect the behavior of $C_{\tau_k}(t|0)$. On the other hand from Eq. (3.1a), the probability density must then obey the following scaling relation

$$P_{A\tau_k}(\omega) = A P_{\tau_k}(\omega A) \qquad (3.3)$$

Consider now N subsystems each of which is characterized by a member of the set of independent and identically distributed stochastic variables ω_i. We want then a total system made up of these subsystems and characterized by a normalized summand variable

$$\omega = B_{Nk}^{-1} \sum_{i=1}^{N} \omega_i \qquad (3.4)$$

such that the probability density for ω is also the Fourier transform of the same k-dominant characteristic function as that for the individual systems. The normalization function B_{Nk} is expected to depend on both N and k. Such a probability density will be called k-dominant stable. We write

$$P_{\tau_k}(\omega) = \int_{-\infty}^{\infty} \ldots \int_{-\infty}^{\infty} \delta\left(B_{Nk}^{-1} \sum_{i=1}^{N} \omega_i - \omega\right) \prod_{i=1}^{N} P_{\tau_k}(\omega_i) d\omega_i \, , \qquad (3.5)$$

which from Eq. (3.3) can also be written in the form

$$P_{\tau_k}(\omega) = B_{Nk}^{N} \int_{-\infty}^{\infty} \ldots \int_{-\infty}^{\infty} \delta\left(\sum_{i=1}^{N} \bar{\omega}_i - \omega\right) \prod_{i=1}^{N} P_{\tau_k}(B_{Nk} \bar{\omega}_i) d\bar{\omega}_i \qquad (3.6)$$

Here

$$\bar{\omega}_i = B_{Nk}^{-1} \omega_i \qquad (3.7)$$

Transforming the δ-function to t-space and carrying out the $N \bar{\omega}_i$ integrals, we obtain

$$P_{\tau_k}(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} [C_{B_{Nk}\tau_k}(t|0)]^N \cos \omega t \, dt \qquad (3.8)$$

The time arguments in the products of $C_{B_{Nk}\tau_k}(t|0)$ in the integrand are the same so the resulting product of N $C_{B_{Nk}\tau_k}(t|0)$'s has the piecewise structure of $C_{B_{Nk}\tau_k}(t|0)$. However, Eq. (3.8) is not a proper equation unless

$$[C_{B_{Nk}\tau_k}(t|0)]^N = C_{\tau_k}(t|0) \qquad (3.9)$$

Using the monomial exponential property of all the segments of $C_{B_{Nk}\tau_k}(t|0)$ and continuity conditions Eqs. (2.3a,b) as appropriate, we find Eq. (3.9) follows if

$$B_{Nk} = N^{1/k} \qquad (3.10)$$

We have the result then that k-dominant stability is achieved for a prob- ability density of a summand variable that is normalized by $N^{1/k}$. For k=2, $N^{1/2}$ provides an enhancement in ω with respect to the ω_i. For k=1, ω and the individual ω_i coincide while for k=α, $0<\alpha<1$, ω is reduced with respect to an individual ω_i.

We can view the normalization as a manifestation of an interaction envi- ronment that is introduced when the independent subsystems are put together to form the total system. However, once the total system is formed, it can be viewed as a building block subsystem for a larger total system. It follows that the interaction environment for the single system must be self-consistent with the interaction environment for the total system. An important question is how the interaction environment is first set up on a microscopic basis or alternatively, if k-dominant stability is taken as a primary principle, how a microscopic interaction environment is arranged to form the appropriate build- ing block. In the next section we consider a model of relaxation in complex systems that can be viewed as providing a microscopic basis for α-dominant behavior. If k-dominant stability is taken as the primary principle, all models of relaxation must be constructed in such a way to yield k-dominant behavior.

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In general, this complementarity of microscopic basis and k-dominant stability may be viewed in the following physical way. Theoretical descriptions of relaxations in a complex physical systems such as glasses and polymers are usually based on a microscopic relaxation species. The latter can e.g. be a dipolar group, a hopping charge, a spin, or an entire polymer chain or segment of it. A particular relaxation species i in a complex environment and interacting with a nominal heat bath, will have behavior described in terms of a a relaxation function $\phi^i(t)$, or a characteristic function $C_{\tau_k}^i(t|0)$ or a probability density $P_{\tau_k}^i(\omega)$. Measurements in relaxation are macroscopic quantities such as transient current, stress, magnetization, volume and shear flow. The stochastic variable ω of the macroscopic quantity is a normalized sum of the form of Eq. (3.4). It is generally assumed in all considerations of relaxation that the macroscopic $\phi(t)$ is identical to the microscopic $\phi^i(t)$. This assumption is equivalent to the assumption that $P_{\tau_k}^i(\omega)$ is a stable probability density or in our description, k-dominant stable. From the results of this section, these equivalent assumptions are consistent with the requirement that $\phi^i(t)$ be the positive time part of a k-dominant $C_{\tau_k}^i(t|0)$ of the form given in Eq. (2.2).

As a final comment in this section, it is interesting to consider the relationship between Lévy stability[14,11] and the k-dominant stability introduced here. While we have restricted ourselves to real characteristic functions that must be piecewise with multiple monomial exponential behaviors, Lévy deals with characteristic functions with magnitude equal to a single monomial exponential with monomial power g, 0<g<2. Except for the possibility of the quadratic exponential behavior, such monolithic behavior cannot meet the physical conditions on the autocorrelation function at t=0. For a real characteristic function, the monolithic quadratic exponential is ruled out by the

physical unrealistic requirement that its instantaneous decay rate is proportional to t and therefore blows up as $t\to\infty$ and such behavior is never observed. For any characteristic function, the limitation to a quadratic exponential magnitude at intermediate times is not sufficiently general to provide a satisfactory basis for the description of decay phenomena. Thus from the viewpoint of physics, the present generalization to k-dominant stability and its concomitant piecewise characteristic function, or some further generalization yet to be made, is a necessary step beyond Lévy stability.

IV. Comparison with the Ngai Model of Relaxation

We now take advantage of the fact that $C_{\tau_k}(t|0)$ for $t>0$ can be identified as a step response function to compare our constructed $C_{\tau_k}(t|0)$ with the relaxation function obtained in the so-called Ngai model of relaxation. The latter model predicts: (1) a fractional exponential decay in a time regime $t\gg\omega_c^{-1}$ when $\omega_c\tau_\alpha\gg1$; (2) a relationship between a relaxation time τ_1 in a linear exponential regime $t\ll\omega_c^{-1}$ and τ_α. Here ω_c is a cutoff frequency that is a parameter of the model as described below and we have taken the liberty of using τ_k, $k=\alpha,1$ to suggest the direct connection with our $C_{\tau_k}(t|0)$. It is clear that the model predictions are consistent with k-dominant behavior for $C_{\tau_k}(t|0)$ and, as we shall see in detail below, the continuity condition Eq. (2.3b). Thus the model may indeed provide insight into the physics of the formation of $C_{\tau_k}(t|0)$ and the question of the microscopic origin of the building blocks of k-dominant stability.

To provide context for such a discussion, we briefly review the concepts in the Ngai model. The model considers a relaxing entity that is coupled to a heat bath on a microscopic level. In accordance with the conventional description of relaxation, after some induction time, say $t>T_Q$, the system initially

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relaxes according to a linear exponential. The complexity of the system is modelled in terms of correlations imposed on the simple system. These correlations are represented by modes which are distinct from the heat bath but which also interact with the relaxing entity. These modes were called correlated state excitations in Ref. 6. The correlated state excitations themselves modell. in terms of the energy level spacings of the Gaussian Orthogonal Ensemble (GOE) of random matrix Hamiltonian theory.[12] The level spacing distribution of the GOE gives the energy density function $N(E)$ of the correlated state excitations which are also called level spacing excitations.[7] $N(E)$ varies linearly with E up to an upper cutoff $E_c = \hbar\omega_c$ that defines the low frequency regime, levels off to a plateau at intermediate energies, and vanishes (linear) exponentially at high energies. As noted above, long and short time regimes for the model are quantified in terms of ω_c, i.e. $\omega_c t \gg 1$ means t in the long time regime while $\omega_c t \ll 1$ means t is in the short time regime (for us, intermediate time regime).

The relaxing entity is then not only coupled to the heat bath but also to the energy level spacing or correlated state excitations. The latter coupling strength is assumed to be independent of the energy of the excitations. It is important to emphasize that the level spacing excitations are not part of the heat bath. The excitations are in equilibrium prior to system excitation, are driven out of equilibrium during the relaxation by their interaction with the measured relaxing entity, and eventually return to equilibrium in a process not addressed specifically in the model.

The interaction between the relaxing entity and the level spacing excitations leads to a long time fractional exponential decay as a manifestation of the modification of the instantaneous decay rate from $W_1(t) = \tau_1^{-1}$ for $\omega_c t \ll 1$

$$W_\alpha(t) = W_1(t) \exp[(\alpha-1)\gamma] (\omega_c t)^{\alpha-1} , \quad 0 < \alpha < 1, \quad \omega_c t \gg 1 . \tag{4.1}$$

Here γ is the Euler constant. Instead of α, the model is usually formulated in terms of the parameter

$$n = 1-\alpha \qquad (4.2)$$

where $n = 2C|V|^2$. $\qquad (4.3)$

Here C is the slope of the linear portion of the density of states N(E) and $|V|^2$ is the averaged squared magnitude of the coupling interaction between the relaxing entity and the level spacing excitations. As mentioned above, $|V|$ is assumed to be independent of the energy of the excitation.

The long time step response function is written as

$$\phi(t) = \exp{-(t/\tau_\alpha)^{1-n}} \quad , \quad \omega_c t \gg 1 \qquad (4.4)$$

It should also be noted that Eq. (4.4) applies for experimental observations where $\omega_c \tau_\alpha \gg 1$ so $\phi(t)$ meets the condition (c) of α-dominance. Given Eq. (4.4), the instantaneous decay rate for $\omega_c t \gg 1$ is

$$W_\alpha(t) = \alpha t^{\alpha-1}/\tau_\alpha^\alpha \qquad (4.5)$$

Consistency of Eqs. (4.1) and (4.5) requires that

$$\tau_\alpha^\alpha = \alpha \exp[(1-\alpha)\gamma] \; \omega_c^{1-\alpha} \; \tau_1 \qquad (4.6)$$

Equations (4.4) and (4.6) are the predictions of the Ngai model which are readily susceptible to experimental verification. For example, let us consider the effect of elevated temperature on relaxations in complex systems such as amorphous polymers and glasses which at normal temperatures have the α-dominant form of Eq. (4.4) and τ_α is thermally activated. The Ngai model has an <u>additional</u> prediction given by Eq. (4.6) which relates τ_α to τ_1. If τ_α has an Arrhenius temperature dependence with activation energy E_a^*, then τ_1 will have a different but predictable activation energy of $E_a \equiv \alpha E_a^*$. More generally, a dependence $f(\zeta)$ of τ_α on a physical parameter ζ, such as molecular weight M of

a polymer chain in polymer melt relaxation or isotope mass m of alkali ion in conductivity relaxation of glasses, will correspond to a different $[f(\zeta)]^\alpha$ dependence on ζ for τ_1. These predictions have been verified in many different complex systems.[14]

An interesting possibility that is allowed in the Ngai model is the cross-over from α-dominant to 1-dominant behavior by changing the conditions of physical measurement. To discuss this point further, we complete the contact between the Ngai model and the construction of $C_{\tau_\alpha}(t|0)$ by identifying T_L in Eq. (2.3b) to be

$$T_L = \alpha^{1/(\alpha-1)} \exp(-\gamma) \omega_c^{-1} \qquad (4.7)$$

Then we can consider a change in physical conditions by which T_L does not vary significantly but where τ_α is changed by many orders of magnitude so that a crossover occurs. In such a case, the new τ_1 can obey

$$\tau_1 \ll T_L \qquad (4.8)$$

and the behavior is 1-dominant because it is now τ_1 that is in the interval (T_Q, T_L). Perhaps surprisingly, such a situation occurs in a straightforward way in the temperature dependence of the relaxation of several materials including the glass-forming fused salt[14] $0.4Ca(NO_3)_2 \cdot (0.6)KNO_3$, the ionic conductor sodium β-alumina,[15] and in polymeric systems.[16]

It may be noted that most presently proposed models[17],[18] of relaxation encompass neither the prediction of Eq. (4.6) nor the possibility of crossover of type of dominant behavior. Although the Ngai model does not require the sudden discontinuity by changing the decay rate from $W_1 = \tau_1^{-1}$ to $W_\alpha(t)$ at the single point $t = T_L$, it is compatible with such an interpretation. Then instead of the more abstract discussion of the temporal analog of thermodynamic phase transitions in section II, we may now consider $W_1(t)$ to be suddenly dressed by

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means of interaction with correlation or level spacing excitations at T_L to yield the decay rate $W_\alpha(t)$.

Correlation excitations may also be viewed as providing the vehicle by which α-dominant stability is introduced microscopically. Thus the Ngai model may be viewed as providing the first building block. On the other hand, as indicated in section III, we may view α-dominant stability as a primary principle. Then the microscopic interactions that enter the first building block must be compatible with α-dominant (or more generally any k-dominant) stability.

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